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Differentiating Special Nuclear Materials through computationally generated gamma-ray spectra

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Abstract

This work focused on providing a first step interpretation of fission product gamma-ray spectra from 0.1 hours to 10 days after a fission event. The activity of fission products from mass chains 89 through 92 and 131 through 141 was evaluated using independent fission yields. An MCNP model was used to simulate the gamma-ray spectra from a single HPGe crystal for the isotopes of interest for a sample of U-235 and Pu-239 assuming a fission neutron spectrum. The ratios of the activities were evaluated for differences in the spectrum. The analysis was optimized for isotopes with prominent gamma-ray branches and a large difference in intensities between U-235 fission versus Pu-239 fission.

Keywords

MCNP, Gamma spectroscopy, SNM, HPGe, Monte Carlo

Introduction

Fission occurs when special nuclear material (SNM), such as U-235 and Pu-239, absorbed a neutron. Fission generates a large amount of energy via the radioactive decays of fission fragments. The fission is either in controlled environment, as in nuclear reactors, or an uncontrollable setting, as in nuclear weapons. Nuclear forensics seeks to

differentiate the fissile materials used in the fission process. One differentiation method is made by measuring the radioactive decay of fission fragments. The distinction between different fissile materials is possible because of the distributions of fission fragments that are dependent on the materials and incident neutron energies.

This paper focuses on simulation studies of Pu-239 and U-235 fission impinged with fission energy neutrons. The interest lies in detecting gamma-rays emitted from decaying fission fragments via high resolution high purity germanium (HPGe) detectors [1]. The model assumes the usage of a single crystal HPGe detection system, which is widely commercially available. Recent advancements in mechanically-cooled HPGe detectors, bypassed the need of liquid nitrogen cooling, enhance the deployability of such detection systems so that early time detections of gamma-ray from fissions can be made. Fission creates a large number of isotopes with half-lives of varying length and complex decay chains. The gamma-ray spectrum of a fission SNM sample is time-dependent and complex with different gamma-ray energies due to different population of fission nuclides present as the sample decays. The spectrum is constantly evolving through the decay of isotopes and the build-up of longer lived parents as the time from fission increases.

This investigation focused on mass chains with volatile or semi-volatile components. The method breaks complicated spectra into simpler parts and provides a jumping off point for diagnostics in a simplistic system. In order to address the possibility of in-situ and off-site samplings, the focus was on mass chains that have an isotope that could leave the origin of the fission event, specifically mass chains that have a krypton or xenon isotope. Due to the complexity of fractionation, geometric and chemical, this paper focuses on the differentiation using the entire sample.

One of the limiting factors of this research is the error associated with the nuclear data, specifically the independent fission yields. The error in the recommended independent fission yields from England and Rider [2] can be over 45% for some of the isotopes used in this analysis. However, with recent focus using critical assemblies at device assembly

facility (DAF) in Nevada, further basic science studies can be made and will result in smaller errors in the determination of fission yields.

Model/Method

This work focused on solving the decay chains from the time of fission to 10 days post-fission. The solutions were used to generate gamma-ray spectra of a sample at several temporal points using MCNP5 [3]. The absolute amount of an isotope produced from the fission event was based on the recommended independent fission yields [2]. The model assumed 1.45×10^{23} nuclei of the specific SNM, U-235 or Pu-239, fissioned. An MCNP5 model was used to simulate the gamma-ray spectrum generated by a single cylindrical germanium crystal. The front face of the crystal is placed at 6.265 cm from the 0.01 cm thick fission product sample. The spectra were generated for two crystal sizes. The first was a 7.5 cm long cylinder with a radius of 2.85 cm. The second crystal size was 1 cm radius with 2 cm length. The former crystal size was used to match up gamma-ray spectra collected with an actual detector in use at the Lawrence Livermore National Laboratory. The later crystal was chosen for the ease of extrapolation to other crystal sizes. Simulated energy dependent count ratios between the large crystal detector and the smaller crystal detector is shown in Fig. 1.

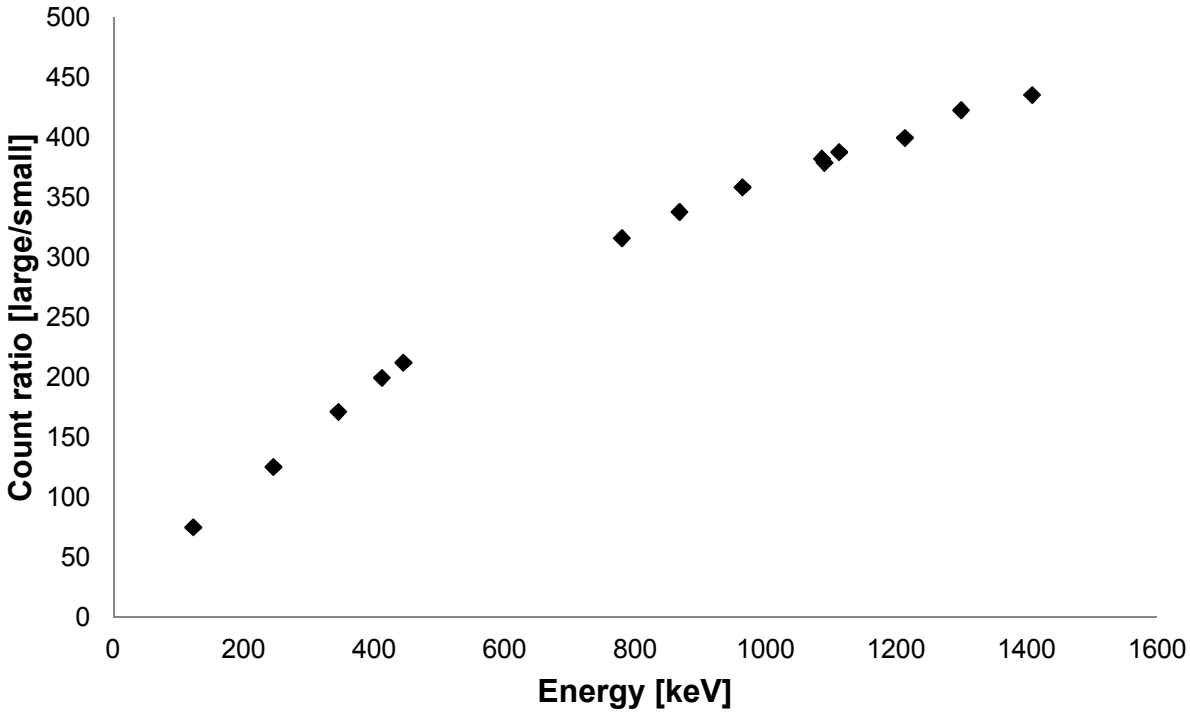
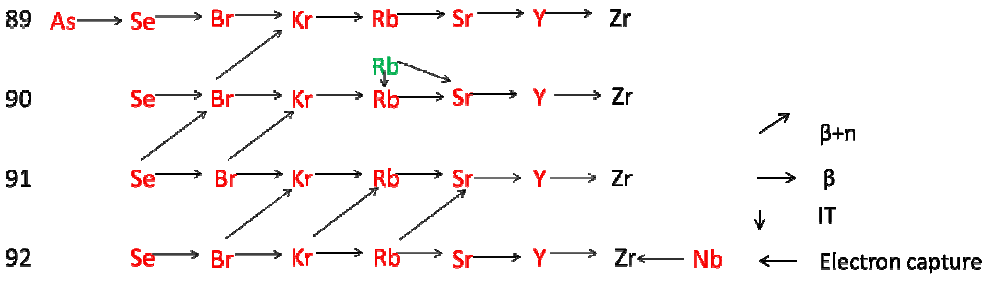


Fig. 1 Simulated gamma-ray energy versus intensity ratios from the two detectors using an Eu-152 source

The ratios of gamma-ray intensities at given energies were used to differentiate between the fissile materials used. Therefore, the work can be scaled with different detector size with background compositions as the limiting factor.

The Bateman equations for decay of mass chains 89 through 92 and 131 through 141, shown in Fig. 2, were generated and solved for using a matrix method and Laplace transforms. Previous work in this area focused radioxenon activities in mass chains 131, 133, and 135 [5,6]. This work also accounts for the build-up and exit from a specific mass chain through β + neutron decays. This correction is small relative to the overarching problem of fission yield data uncertainty, so previous group ignored the effect. The mass chains were chosen to the presence of a noble gas component, xenon or krypton, in the mass chain. The equations were input into a matrix for a solution [9]. The solution for each mass chain was used to provide isotope activities from time frames from 0.1 hour to 10 days post-fission. The ratio of isotopic activity to total activity is

shown in Fig. 3 and Fig. 4. There are noticeable differences between the Pu-239 (Fig. 3) and U-235 (Fig. 4) fission products.



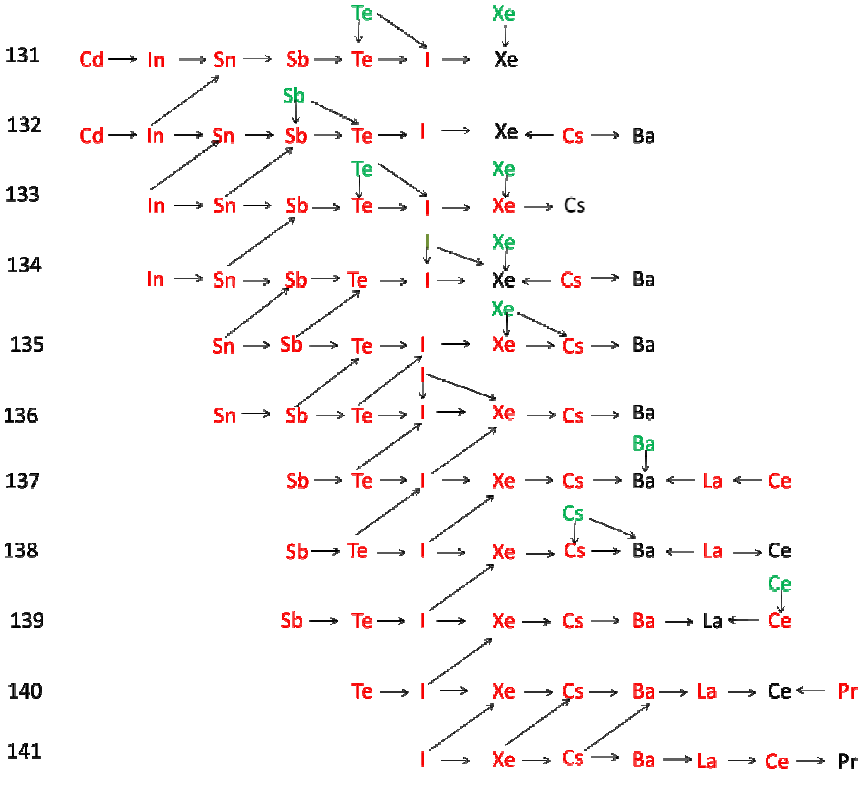


Fig. 2 The decay chains included in the model. The number on the right is the mass chain. Green represents a metastable state, black are stable isotopes, and red is the radioactive ground state of an isotope.

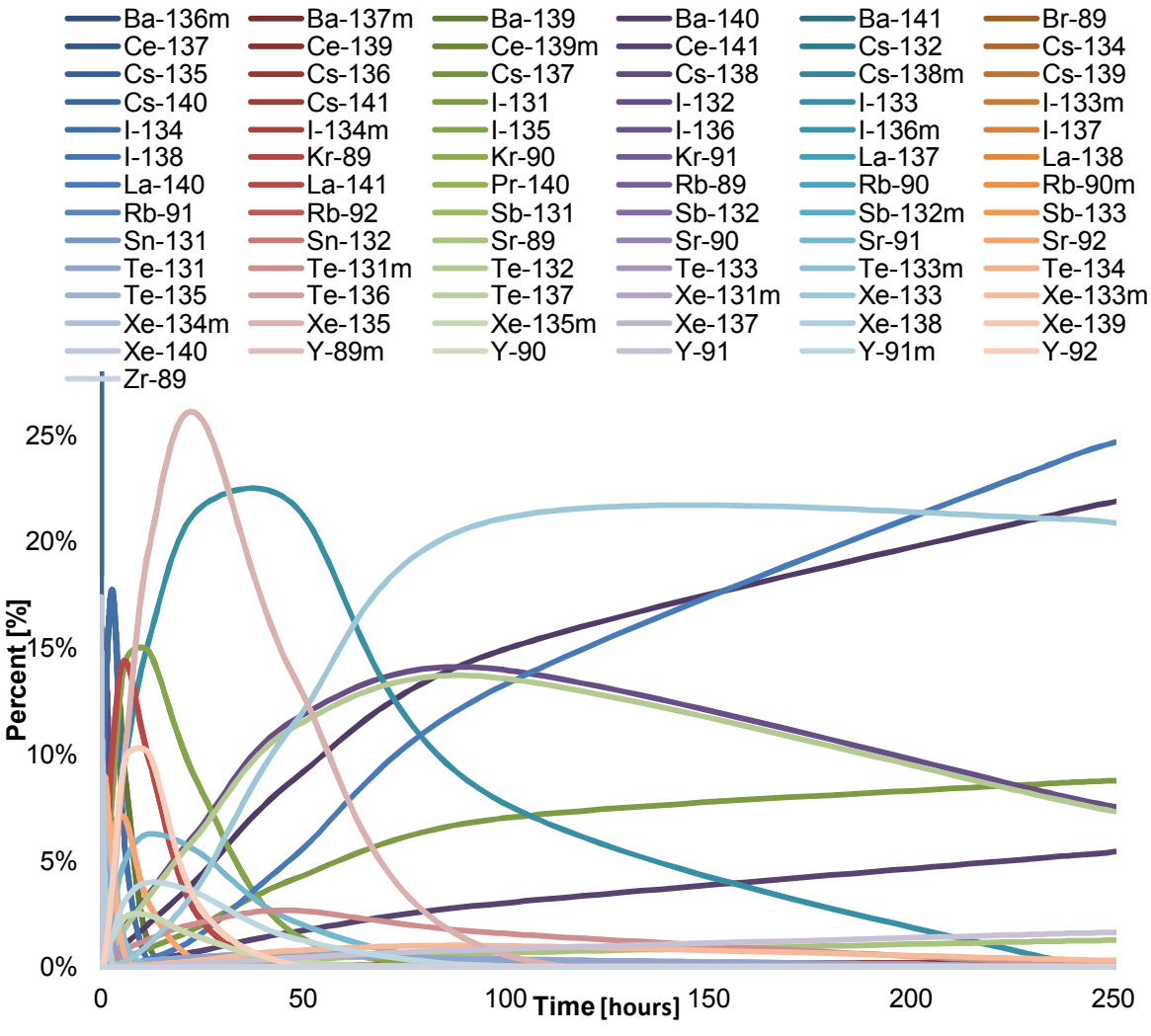


Fig. 3 The percentage of isotopic activity to total activity of the sample for Pu-239 fission with fission spectrum energy neutrons

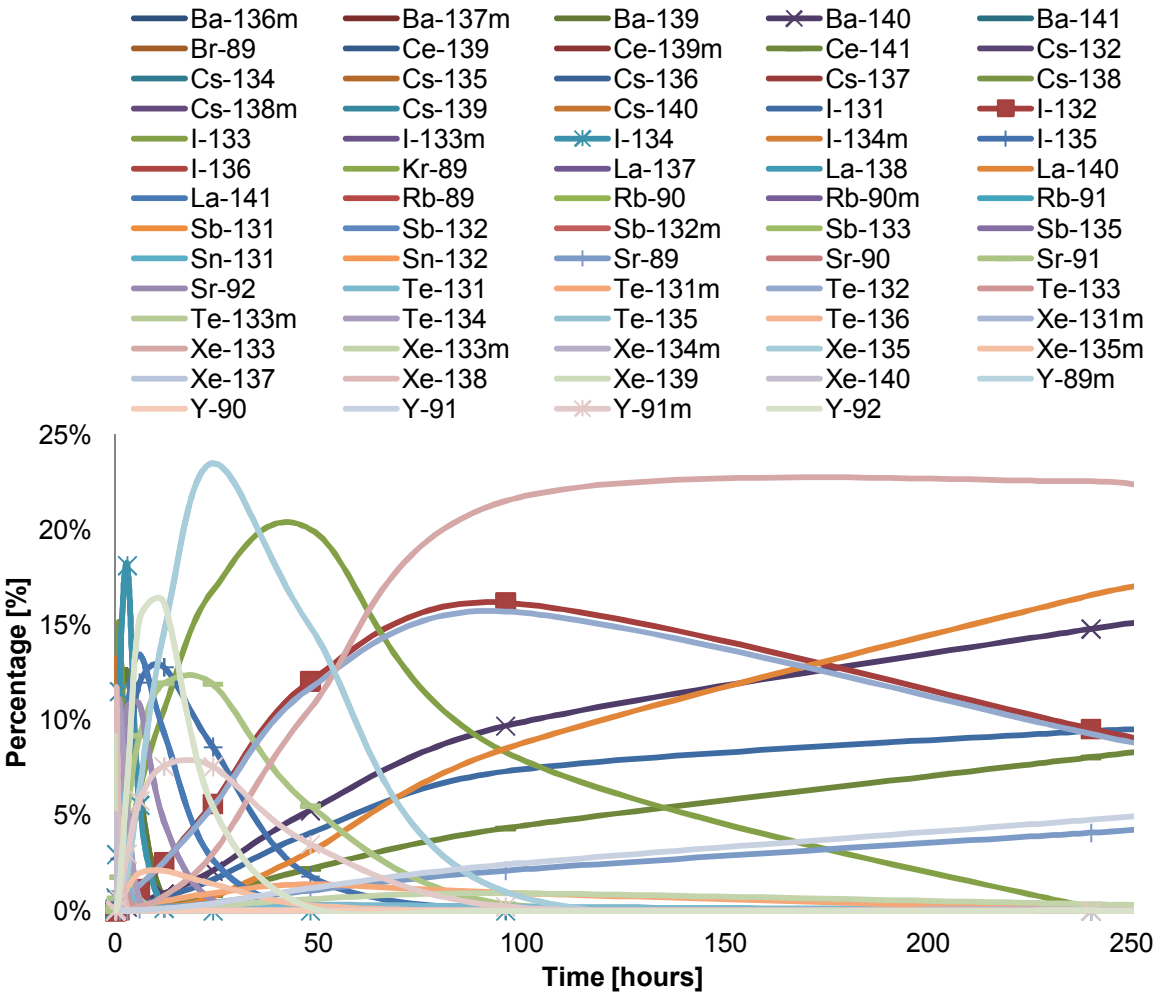


Fig. 4 The percentage of isotopic activity to total activity of the sample for U-235 fission with fission spectrum energy neutrons

Simulated Gamma-ray Spectra with HPGe Detectors

The HPGe gamma-ray spectra were generated using an MCNP5 model of the fission products at a specific decay time. The amount of the isotope and the intensity of the gamma-rays from the isotope decay [10] for each of the decay pathways were input into the model. The mass chains of interest gamma-ray spectra for U-235 and Pu-239 one day post-fission using the larger, realistic HPGe crystal size is shown in Fig. 5. The spectra

are similar, several gamma-ray energy peaks show dramatic change in the intensity that could be used to differentiate fissile material used. There are situations where the fission time will be unknown. Therefore, the decay time will be unknown. The solutions sought out are independent of decay time. This requires the differences in early activity and in-growth to stay relatively constant.

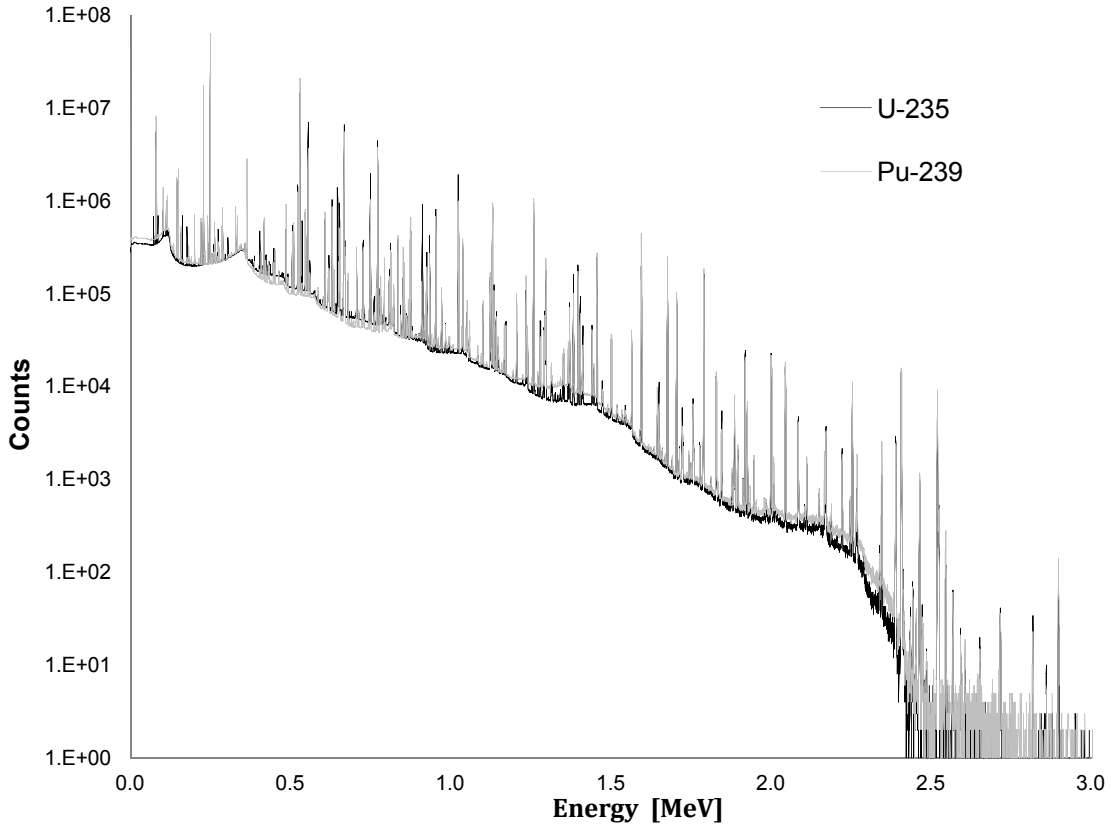


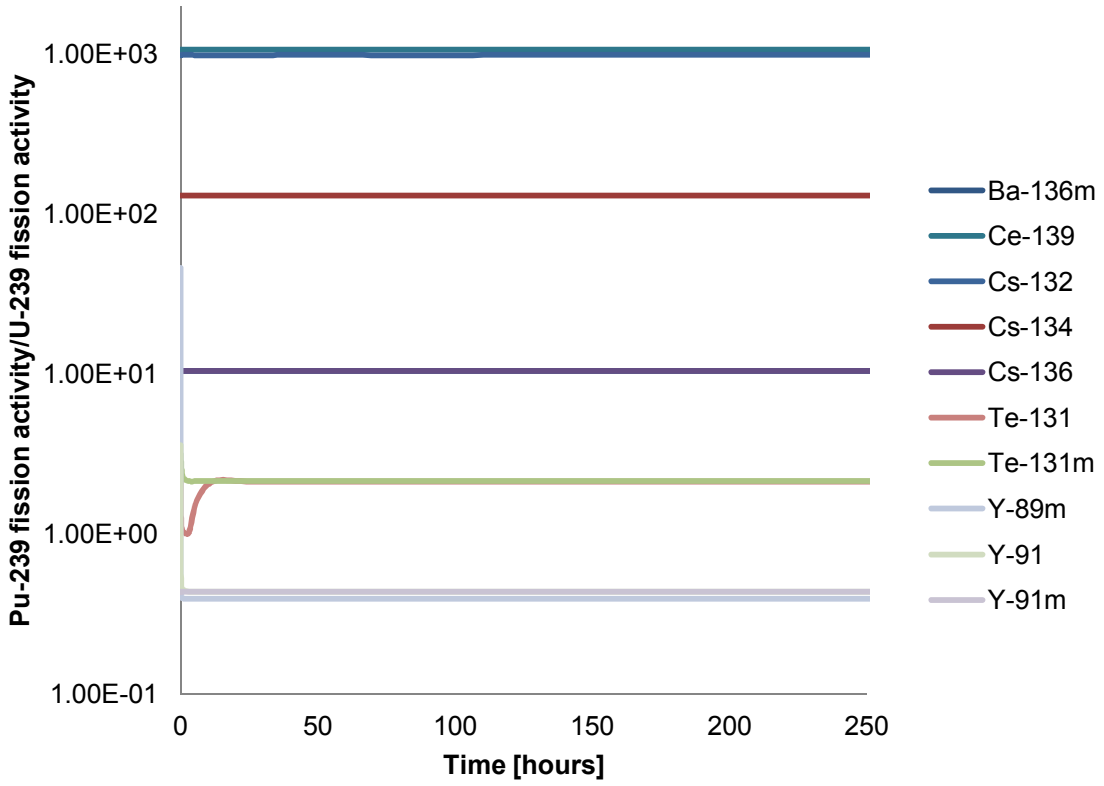
Fig. 5 Simulated gamma-ray spectra from the Pu-239 and U-235 fission with 1 day of decay time. The HPGe crystal cylinder has a dimension of 7.5 cm in lengthand 2.85 cm in radius

Since it will be impossible to capture all of the fission products to calculate the total activity produced of any single isotope, it is useful to use isotopic ratios. If we rely on the assumption that all isotopes escaped at the same frequency (completely intermixed),

we can use ratios of isotopes from various fission chains. This analysis also focuses on ratios with at least a 2:1 production difference due to the large errors currently associated with the fission yields database for these mass chains, shown in Table 1. The exception is La-140 and Ba-140 with ratios that settle near 1.9:1 and are used readily by the nuclear community in fission event analysis. Fig. 6 shows ratios of the isotopes that allow the differentiation between U-235 fission and Pu-239 fission.

Table 1 Radioisotopes with fission yield production differences

Isotope of Interest	Energy [10] of strong gamma-ray lines [keV]	
Ba-136m	818.514	1048.073
Ba-140	537.261	
Ce-139	165.853	
Cs-132	667.718	
Cs-134	604.721	
Cs-136	818.517	1048.073
La-140	1596.21	
Te-131	452.323	1146.96
Te-131m	773.67	852.21
Y-89m	908.960	
Y-91	1204.77	
Y-91m	555.57	



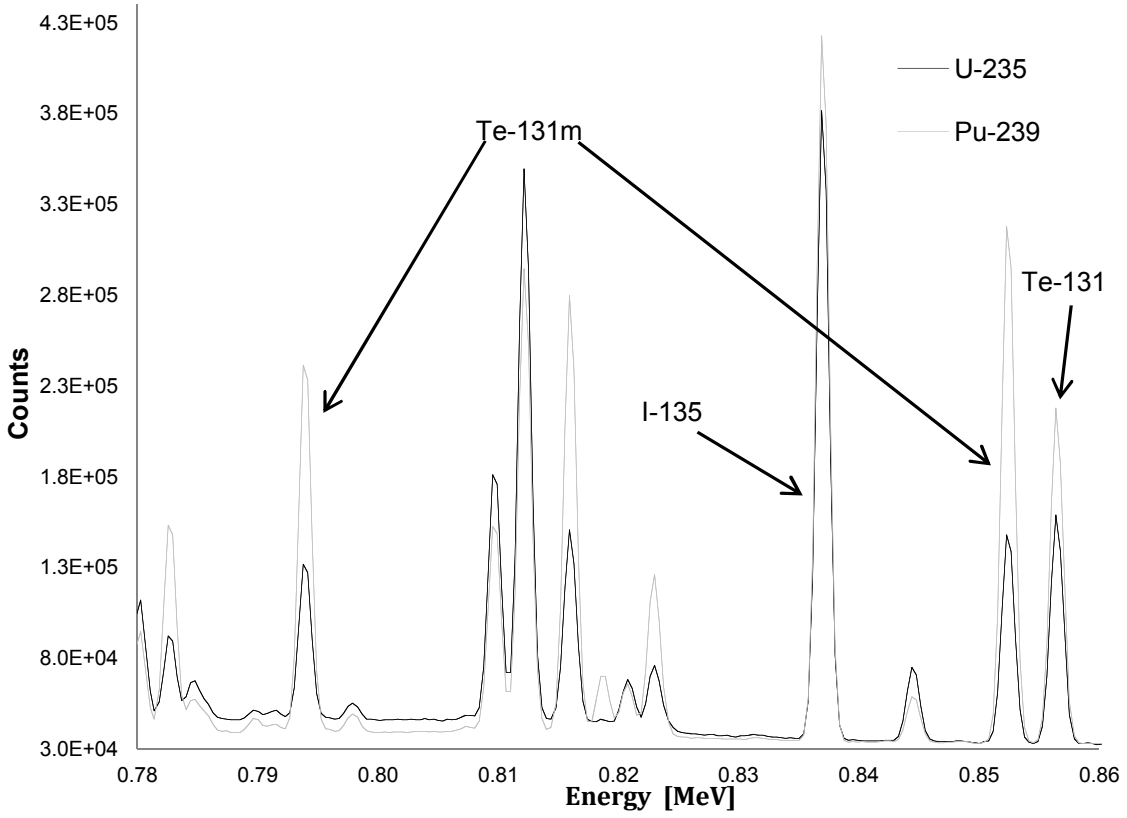
131

132 **Fig. 6** The isotope ratios versus time for Pu-239 to U-235

133 Isotopic ratios between the fission products produced from U-235 or Pu-239 fissile
134 material with fission energy neutrons show many of the isotopes in these mass chains we
135 selected converging to 1, which would deem the fission of the different fissile material
136 types indistinguishable. One of the most differentiable ratios is for Zr-89. This is due to
137 a small amount created in Pu-239 fission with an associated error of the independent
138 fission yield over 45%, but none produced in the U-235 fission. The small amount would
139 probably be undetectable with only 1.68×10^{-21} atoms of Zr-89 produced per fission. Zr-
140 89 will be left out of further discussion due to the uncertainty, small yield and possible
141 incomplete nuclear data set.

The Te-131m ratio is steadily around 2:1 before 1 hour post-fission event. Te-131 builds into a ratio of approximately 2:1 after 12 hours. Both are fueled by the fission yield of Sb-131, half-life 23.0 min, being significantly larger for Pu-239 than U-235. Cs-132 also has a significant difference due to differences in very small, not well-known fission yield for both U-235 and Pu-239. Due to the small amount produced, Cs-132 would be difficult to detect. However, Cs-132 has a strong gamma line at 667.718 keV for its primary mode of decay, electron capture to Xe-132. Cs-136 has several strong gamma-ray lines, shown in Table 1, and a production ratio above 10. Cs-134 has a Pu-239 to U-235 production ratio of 131.

The 131 mass chain provides an intertwined relationship between I-131, Te-131, and Te-131m. The Te-131m/I-131 ratio may be the most consistent. However, the Te-131/I-131 ratio may be the relationship to consider due to the higher activity of Te-131 and higher intensity gamma-ray lines—149.716 keV, 773.67 keV, 852.21 keV, 793.75 keV, and 1125.46 keV—in various points along the spectrum. The peaks between 780 keV and 860 keV are shown in Fig. 7.



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158 **Fig. 7** Simulated gamma-ray energy spectra at 1 day decay time, only with energy
159 between 780-keV and 860-keV were plotted for comparison. Differences in peak areas
160 are shown in the figure. The peaks highlighted in the figure are mostly comprised of
161 gamma-rays from a single mode of radioactive decay from a single isotope.

162 From the smaller mass chain cluster, yttrium has strong gamma-ray lines and a shifted
163 Pu-239 to U-235 fission yield. The larger mass chains focused on more production by
164 Pu-239 fission while the smaller mass chains have a larger production from U-235
165 fission. Y-89m develops to a 0.395 Pu-239 to U-235 production ratio at an hour. Y-91
166 becomes a steady 0.439 ratio at 3 hours. Y-91m provides a steady ratio as early as 18
167 minutes post-fission with a 0.438 production ratio.

168 **Error**

The independent fission yield error is the overwhelming contributor to the uncertainty calculation of this project. While the cumulative fission yield data has smaller associated error, the uncertainty calculation is still overwhelmed by this problem.

The other sources of error in the system were minor in comparison. The half-lives, decay pathways, gamma-ray energies, and gamma-ray intensities are other sources of nuclear data error. However, the error is dwarfed by the fission yield error, both independent and cumulative. Another source of uncertainty was the statistical error introduced by MCNP. This error can be reduced with longer run times. In a real world situation, the spectra could be improved with better background quantification and longer count times. None of these steps are helpful with the fission yield uncertainties frequently over 20% error.

The improvement in this method will not come from longer count times. The improvement will come from experimental work quantifying the fission yields, both independent and cumulative, for Pu-239 and U-235 for fission spectrum neutrons [11].

Conclusion

This work does not consider the chemistry effects as in a real world situation, such as a reactor release event. The focus is to determine how useful gamma-ray spectra can be at differentiating a fissile material if we assume a perfect fission energy distribution of the neutron source and total release of fission chains. The research focused on the mass chains with fission products and other volatile or semi-volatile isotopes present, which have a larger probability of escaping the point of fission.

Future work will be focused on expanding to other incident neutron energies and to fissionable material. The data analysis method will also be useful to analyze the gamma-ray spectra associated with different phases in the commercial nuclear power reactor life phase. The phase would include a commercial nuclear reactor fueled with fresh UO_2 fuel, spent fuel ponds, and various points in the nuclear fuel cycle.

Acknowledgements

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